Reply to comments on "Simulating ozone dry deposition at a boreal forest with a multi-layer canopy deposition model"

October 6, 2016

We thank the reviewer's thoughtful comments which are helpful not only for this manuscript but also for our future research. Our reply for all the comments are shown below.

1. **Comments: 1. However, I would have appreciated a more extended parameterization and a better description of the model in order to clearly understand the formalism adopted to predict energy balance terms.**

   We added more details about the energy balance terms, including sensible and latent heat fluxes, soil heat flux and radiation.

   "In SOSAA, the horizontal wind velocity (u and v), temperature (T), specific humidity (q_v), turbulent kinetic energy (TKE) and the specific dissipation of TKE (ω) are computed every time step (10 s) by prognostic equations. In order to represent the local to synoptic scale effects, u, v, T and q_v near and within the canopy are nudged to local measurement data at SMEAR II station with a nudging factor of 0.01. A TKE-ω parameterization scheme is used to calculate the turbulent diffusion coefficients (K_t) (Sogachev, 2009),

   $$K_t = C_\mu \frac{\mathrm{TKE}}{\omega} \tag{1}$$

   $$\omega = \frac{\varepsilon}{\mathrm{TKE}} \tag{2}$$

   where ε is the dissipation rate of TKE and C_μ is a closure constant. Hence the turbelent flux of a quantity X ($F_{t,X}$) can be computed as

   $$F_{t,X} = -K_t \frac{\partial X}{\partial z} \tag{3}$$

   where upward fluxes are positive and vice versa. Specifically, the sensible heat flux (H) and latent heat flux (LE) at each model layer are computed as

   $$\mathrm{H} = -C_{p,air}\rho_{air}K_t\left(\frac{\partial T}{\partial z} + \gamma_d\right) \tag{4}$$

   $$\mathrm{LE} = -L_v K_t \frac{\partial q_v}{\partial z} \tag{5}$$

   where $C_{p,air}$ (1009.0 J kg^{-1} K^{-1}) is the specific heat capacity at constant pressure. ρ_{air} (1.205 kg m^{-3}) is the air density which is a constant in the model. γ_d (0.0098 K m^{-1}) is the lapse rate of dry air. L_v (2.256×10^6 J kg^{-1}) is the latent heat of vaporization for water."

   "The upper boundary values of u, v, T and q_v are constrained by the ERA-Interim reanalysis dataset provided by the European Centre for Medium-Range Weather Forecasts (ECMWF, Dee et al., 2011). At the canopy top, the incoming direct and diffuse global radiations measured

Table 1: The average and standard deviation of modelled and measured (OBS) O_3 fluxes above the canopy for different conditions in different cases are shown. The relative change of modelled O_3 flux compared to the observation $(F_{t,mod} - F_{t,obs})/F_{t,obs}$ is also listed within the parentheses.

cases	ALL	D	N
OBS	0.125 ± 0.090	0.171 ± 0.085	0.052 ± 0.037
RSOIL200	0.146 ± 0.090 (+16.6%)	0.192 ± 0.085 (+12.3%)	0.070 ± 0.034 (+34.9%)
BASE	0.128 ± 0.079 (+1.93%)	0.168 ± 0.075 (-1.51%)	0.061 ± 0.030 (+16.1%)
RSOIL600	0.118 ± 0.075 (-5.85%)	0.156 ± 0.070 (-8.64%)	0.055 ± 0.029 (+5.07%)
RSOIL800	0.112 ± 0.072 (-10.7%)	0.148 ± 0.067 (-13.0%)	0.051 ± 0.028 (-2.28%)

at SMEAR II station, and the long wave radiation obtained from the ERA-Interim dataset are read in to improve the energy balance closure. Then the reflection, absorption, penetration and emission of three bands of radiation (long-wave, near-infrared and PAR) at each layer inside the canopy are explicitly computed according to the radiation scheme proposed by Sogachev et al. (2002). At the lower boundary, the measured soil heat flux at SMEAR II are used to further improve the representation of surface energy balance. All the input data are interpolated to match the model time for each time step. With the input data, the mass and energy exchange between atmosphere and plant cover (including the soil underneath) and the radiation attenuation inside the canopy are optimal to simulate the micrometeorological drivers of O_3 deposition at this site."

2. **Comments: 2. There are some arbitrary choices of parameters, and not a convincing analysis of sensitivity or results from a model calibration. A table showing results from a sensitivity analysis should be provided.**

   We added a sensitivity test of r_{soil} as below:

   "r_{soil} varied in different studies, ranging from 10 to 180 s m^{-1} for dry soil and 180 to 1100 s m^{-1} for wet soil (Massman, 2004). In this study the dry deposition module was developed on the basis of the model from Ganzeveld and Lelieveld (1995) in which r_{soil} is 400 s m^{-1}. In order to assess the uncertainties involved in estimating r_{soil}, different values of r_{soil} ranging from 200 to 800 s m^{-1} were tested in this study (Table 1). As can be expected, the modelled O_3 fluxes decreased as r_{soil} increased. The BASE case showed the best performance in general, although it overestimated $\sim$ 16% nighttime O_3 fluxes. Since the RSOIL200 case overestimated O_3 fluxes by $\sim$ 17% in average for the whole month, $\sim$ 12% at daytime and $\sim$ 35% at nighttime, the RSOIL200 sensitivity case indicates that using this lower estimate, a value that might be more appropriate for high organic (and dry) soils, seems to not properly represent the role of soil removal at this site. On the other hand, taking higher resistance values, e.g., one of 600 or 800 s m-1 seems to result in a better simulation of the role of the soil uptake at nighttime. However, considering the overall performance and better estimation of daytime O_3 fluxes, we still use 400 s m^{-1} as the soil resistance."

3. **Comments: 3. Basic questions like: what could be the effect of an increase in air temperature and precipitation regimes on ozone deposition? Are not resolved, although it would have been nice triggering the model for some predictions of Ozone deposition under future environmental changes. In general the paper lacks of more mechanistic explanations of the results, with more discussion on the possible drivers of dry and wet ozone deposition.**

   The reviewer has a point also since we have indicated that the observational dataset included data that were potentially resembling more common future conditions at this boreal forest site. However, in the present study we decided to limit ourselves to analyse the model performance for the contrasting day and night time, wet and dry conditions to evaluate the role of the various substrates in the overall O_3 removal. This also reveals the potential significance of non-stomatal removal mechanisms at this site which calls for a better representation of these processes. Such a further improved model could then be applied in follow-up studies to assess what future climate change conditions could imply for removal of pollutants such as O_3 but also other related compounds over boreal forests.

4. **Comments: 4. Pag 2 line 25: You mention again that dew on leaves can increase deposition, but could you spend two lines mensioning the reasons or hypothesis why a hydrofobic molecul reacts so fast on wet surfaces?**

   We added this description in the introduction:

   "Previous studies showed that both the micro structure of the leaf surface and the hydrophilic compounds existing on the leaf surface are able to facilitate the formation of the water films or clusters, although the foliage surface itself is hydrophobic (Altimir et al., 2006). As a result, the different dissolved compounds like organics in the solution formed on leaf surface could react with O_3 and thus enhance the O_3 uptake (Altimir et al., 2006)."

5. **Comments: 5. Pag 3 line 10. What about NOx emitted from soils? Couldn't fast reactions between O3 and NO lead to high O3 fluxes in the sub-canopy region?**

   In SMEAR II station, NO emission is about 6 ng(N) m^{-2} h^{-1} which is close to the detection limit (Pilegaard et al., 2006). Moreover, according to the results in Rannik et al. (2009), the O_3 uptake due to reaction with NO emission is only about 0.0025% (10^{-4} nmol m^{-2} s^{-1} / 4 nmol m^{-2} s^{-1}) of the total nighttime O_3 flux. The sub-canopy O_3 flux at nighttime was about 25–30% of total O_3 uptake, so the effect of reaction with NO on sub-canopy O_3 flux can be ignored.

6. **Comments: 6. Pag 3 line 34: Only one month to test the model? The relative contributions of O3 sinks changes a lot during the seasons in repsonse to air temperature and plant phenology. It is a pity that such an important modelling effort is limited to one month, I would extend to the all vegetative season.**

   It would indeed be nice to conduct an analysis of a full seasonal cycle but this month was giving access to a complete dataset giving the best constraints for the presented detailed evaluation of the model also having still quite some large contrasts. Moreover, first assessing a proper representation of the main drivers of O_3 exchange would then also allow use of the model for full seasonal cycle studies in future research.

7. **Comments: 7. Pag 5 line 5: Extensive research has been conducted in Yuttiala to refine turbulence limitation to flux measurements. Why should we expect an ustar threshold different from other scalars measured at the site?**

   Different scalars may be differently affected by the nighttime phenomena such as accumulation, vertical as well as horizontal advection and in more general by stability conditions. This is due to build up of the concentration gradient which is expected to be particularly large for emitted compounds such as carbon dioxide. Ozone is instead deposited and therefore no large concentration gradients can form, meaning also that the mass balance components other than vertical transport are expected to be smaller. We use the criterion velocity threshold well justified for O_3 e.g. by Rannik et al. (2009).

8. **Comments: 8. Pag 6 line 20: do you have experience of subcanopy O3 fluxes so that you can better parameterize soil reisstances? It seems here that usage of one value rather than another is arbitrary and not properly calibrated.**

   The process of O_3 uptake by soil includes understorey transport (r_{ac}), diffusion at the soil/litter layer interface (r_{bs}) and, finally uptake by this soil/litter layer (r_{soil}) which might be strongly affected by wetness. In this study we ignored r_{ac} since the height of the lowest layer is only about 0.3 m above the ground where vertical transport is mainly limited by the molecular diffusion above the surface which is represented by r_{bs}. r_{bs} will be added in the revised manuscript as:

"The r_{bs} is the soil boundary layer resistance which is calculated as (Nemitz et al., 2000),

$$r_{bs} = \frac{\mathrm{Sc} - \ln(\delta_0/z_*)}{\kappa u_{*g}} \tag{6}$$

Here Sc (1.07) is the Schmidt number for O_3. κ is the von Kármán constant (0.41). $\delta_0 = D_{O_3}/(\kappa u_{*g})$ is the height above ground where the molecular diffusivity is equal to turbulent eddy diffusivity. z_* (0.1 m) is the height under which the logarithmic wind profile is assumed. u_{*g} is the friction velocity near the ground."

For the soil/litter layer resistance r_{soil}, we are aware that application of the value 400 s m^{-1} deemed to represent the global mean soil uptake effciency and is thus a very crude simplication. However, from the conducted sensitivity analysis it can be inferred that this crude representation appears to result in the best representation of both O_3 deposition fluxes as well as O_3 concentration profiles inside the canopy. Actual confirmation of the correctness of the selected value can only be done conducting more detailed soil uptake measurements. Our study also clearly demonstrates the need for such additional measurements.

9. **Comments: 9. Pag 7 line 15. So you mean that Kt has been estimated form measured fluxes? Or in which other way? Reading through the manuscript I feel like the description of the model is not accurate, and more informations should be provided.**

   We added more detailed description about the model SOSAA as described above. So K_t is calculated in the model from a TKE-ω scheme.

10. **Comments: 10. Pag 19 line 15: Can you say that NOx are also not relevant in the boreal forest?**

    Yes, from previous studies, we can conclude that NOx is not relevant to the O_3 uptake in SMEAR II station as we discussed above: At the SMEAR II station, NO emission is close to the detection limit (Pilegaard et al., 2006) and the O_3 uptake due to reaction with NO can be ignored (Rannik et al., 2009).

11. **Comments: 11. Pag 20 line 11: Since the Stomatal resistance is calculated based on evapotranspiration, are you sure that relevant nocturnal soil evaporation does not contribute significantly to Rc? Have you tried to separate canopy transpiration form soil evaporation in the model?**

    Actaully, the stomatal resistance is calculated based on the evapotranspiration from leaves and is already separated from soil evaporation in the model. Therefore, the soil evaporation does not contribute to stomatal conductance in the model.

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